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Translucent Oxides

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Prepared by W. J. GARDNER, J. D. McCLELLAND, and J. H. RICHARDSON

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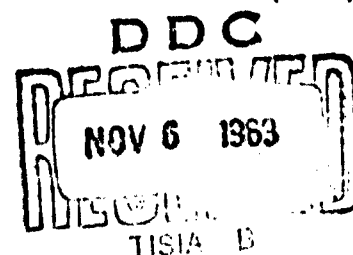
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**AEROSPACE CORPORATION
El Segundo, California**

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This technical documentary report has been reviewed and is approved for publication and dissemination. The conclusions and findings contained herein do not necessarily represent an official Air Force position.

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ABSTRACT

Pure oxide powders of aluminum, beryllium, and magnesium were formed into dense translucent compact bodies by hot pressing. The densities of the bodies were found to be more than 98 percent of the theoretical for the MgO , and more than 99.5 percent for the Al_2O_3 and the BeO . Thermal expansion was measured with a Leitz dilatometer. The data are in agreement with previous published values. Thermal conductivity, modulus of rupture, elastic properties, Young's modulus, shear modulus, dielectric constants, and transmission spectra measurements also are presented.

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I. INTRODUCTION

Recent advances in ceramic fabrication techniques have yielded translucent bodies of alumina, beryllia, and magnesia. Hot pressing has been successfully used for all three materials (Refs. 1-3) while sintering has been used successfully for alumina (Ref. 4,5). The term "Translucent Oxides" has come, in common parlance, to mean bodies the densities of which approach that of a single crystal and which transmit visible light. The properties of these translucent bodies can differ in several ways from those of lesser density. The ability to transmit radiant energy will, of course, change some of the optical properties such as transmittance and might also change the thermal conductivity at high temperature due to a radiant heat transfer mechanism. The lack of porosity, which can be considered as a second phase, might also change the thermal conductivity and the dielectric constant. Finally, those properties which are structure sensitive, particularly the mechanical properties, could be expected to change quite dramatically.

The translucent oxides are new materials in the sense that complete physical, thermal, and optical property data are not yet available; hence, the applications for these materials have not yet been firmly established. However, active interest in these materials has been shown by companies in the electronic, space-aeronautics, and nuclear fields. In the electronic industry, potential applications are seen in klystron tubes, power tubes, antennae windows, and other elements requiring power transmission through an electrical insulator. The expected increases in dielectric strength and resistivity as related to increased density adds to the interest in translucent beryllia, magnesia, and alumina.

The optical properties of the oxides with high transparency in the ultra violet, visible, and near infrared (0.4 to $\sim 9\mu$) are of interest for high temperature instrumentation application because of the thermal stability and oxidation resistance of these materials. Potentially, their economic

value as a replacement for single crystal optical components (lenses, windows, filters, etc.) is significant, especially in the far infrared beyond 50μ .

There is great interest in ceramic oxides as materials for rocket nozzles requiring stop-start capability. This application would take advantage of the minimum oxidation characteristics and chemical inertness of the oxides. Erosion resistance should be improved by the high density of the translucent materials. However, the development of special techniques, such as pre-stressing, are required to provide adequate thermal shock resistance. Such a development is also needed with the nose cap and leading edge application, such as on Dynasoar, for which the general enhancement of mechanical properties achieved through the development of high density oxides may be useful. In those instances where a heat sink is needed, the expected higher thermal conductivity combined with high heat capacity makes beryllia of outstanding interest.

The resistance of beryllia to hot gases and liquid metals is another outstanding property which should be enhanced by translucency because of high density and minimal grain boundary porosity. This combined with low thermal neutron absorption cross section makes beryllia particularly interesting for the nuclear industry.

The high density, fine grain structure of the translucent oxides suggests also general applications such as rotary bearing surfaces, laboratory devices, chemical process windows, radiation windows, special purpose laboratory ware, and vacuum or gas-tight ceramic seals.

The purpose of this report is to describe the fabrication techniques used in preparing translucent oxides by hot pressing and to present available data on microstructure and on thermal, mechanical, and optical properties of these materials.

II. FABRICATION

A. EQUIPMENT

The hot press which was used to fabricate the specimens is shown in Fig. 1. It consisted of a graphite susceptor tube, 8 in. OD, 6 in. ID, and 54 in. in length, placed within a transite shell 16 in. in diameter. The annulus between the susceptor tube and the shell was filled with lampblack which provided thermal insulation. The ends of this annulus were covered with water-cooled brass plates. The susceptor was surrounded by a water-cooled copper induction coil on the outside of the transite shell. Power was supplied by a 3 kilocycle motor generator (Tocco Model G4-4570-01). The furnace assembly was mounted in a steel frame 40 in. in height and 14 ft in length. Two 5-in. hydraulic rams with a travel of 18 in. were placed at the ends of this frame. Pressure as high as 3000 psi to the rams was supplied by means of a hydraulic system. The pressure on the graphite plungers was measured by a precision laboratory pressure gauge placed in the oil line to the hydraulic rams. The pressure system was calibrated in situ by means of a load cell certified by the National Bureau of Standards. Pressure readings had an estimated experimental accuracy of ± 0.5 percent.

The temperature of the die body was measured by means of a disappearing filament optical pyrometer. A graphite tube which extended through the transite case, the lampblack, and the susceptor tube provided access to the die case. The entire temperature measurement system was checked by comparing the pyrometer readings at temperatures below 1500°C with those of a platinum-rhodium thermocouple inserted in the center of a blank die. Excellent agreement was found between the two readings when pyrometer and window corrections were made. Temperature readings were estimated to have a precision of $\pm 10^{\circ}\text{C}$.

The graphite die material was a premium quality, small grain, and relatively flawless graphite designated ATJ Grade. This graphite was chosen primarily

because it could be machined to close tolerance and fine surface finish as well as for its excellent high temperature strength. The dies were baked at a temperature of 1800°C for 4 hr before use to prevent contamination of the compact by outgassing impurities. After the bakeout period, the dies were polished with a cotton flannel cloth to remove any baked-out impurities that adhered to the surface as well as loose carbon.

B. STARTING MATERIALS

The powder used for the fabrication of the translucent beryllia, obtained from the Beryllium Corporation, was High Purity Grade 1. This oxide was prepared by means of a fluoride extraction process that yields a fluoride contamination of approximately three quarters of one percent. The as-received powder was blended and wet screened prior to hot pressing. The starting material for alumina was Linde Type B polishing alumina. Purity of the powder was stated to be greater than 99.9 percent and the particle size was of the order of 0.05μ . The magnesia was fabricated from Baker's AR grade MgO .

C. PRESSING TECHNIQUE

A sufficient quantity of powder was weighed to provide a finished sample that measured $1/4$ in. in thickness and 2 in. in diameter. This powder was poured into a die which had one plunger partially inserted. Care was taken to level the powder because small changes in powder level can produce large density variations in the finished compact sample. For normal specimens such variations are virtually unnoticed because the entire sample is opaque. In translucent specimens, variations of only a fraction of one percent will produce mottled specimens. The second plunger was then inserted into the loaded die and the die assembly was placed in the center of the susceptor tube. Carbon plungers were placed on each end which transmitted the pressure from the rams to the die assembly. The use of carbon reduced heat losses from the hot zone. A sustaining pressure of approximately 200 psi was then applied to the die assembly.

The pressing cycle used for beryllia was as follows: the temperature of the sample was brought to 1750°C and held constant for 10 min to permit the interior of the compact to reach equilibrium. Failure to do this produced a lens-shaped region of non-translucency in the interior of the sample body. The pressure on the sample was then slowly applied in increments of 300 psi until the final pressure of 4000 psi was reached. Ram travel essentially stopped during successive applications of pressure. The sample was held at hot pressing temperature for 4 hr and then permitted to cool overnight.

The temperature-pressure cycle for both the magnesia and alumina differed from that for the beryllia. These former powders began to sinter before the optimum hot pressing temperature was reached. Hot pressing of this partially densified body did not produce the high density bodies which were desired. For this reason pressure was applied to the specimen during the heating cycle. With alumina the restraining pressure of 100 psi was maintained until a temperature of 1000°C was reached. The pressure was then increased gradually until a pressure of 6000 psi and a temperature of 1500°C were reached. For magnesia, the pressure was initially applied at 900°C . The final pressure was 6000 psi and the temperature was 1400°C . In both cases the final pressure was held for 4 hr and the sample was permitted to cool overnight.

The samples as removed from the die were covered with a black coating of carbon which could be removed either by machining or by air firing at 1100°C . The magnesia specimens showed a much deeper penetration than the other powders which indicated a much more severe reaction with the graphite and/or the atmosphere. In some cases large areas of carbon contamination could be seen in the translucent magnesia. This contamination could be explained by reduction of carbon dioxide from the magnesia carbonate. This carbon could be burned out at 1100°C in air leaving a white semitranslucent specimen. The beryllia and alumina did not show such areas of carbon contamination and heating these samples to 1100°C in air did not affect them in any visible manner.

III. PROPERTY DATA

A. DENSITY

When the oxides are hot pressed to high density, there is a tendency for them to become grey in color.^{1,6} This color has often been taken to indicate the presence of colloidal graphite caused by diffusion from the graphite dies at high temperatures. Petrographic examination of these materials, however, shows that the grey areas are regions of transparency. The grey color is probably due to the internal scattering of the incident visible radiation and subsequent absorption within the mass of the sample. It is this lack of reflected light rather than carbon contamination that is the predominant cause of greyness. Dark opaque regions which are not translucent, however, do occur occasionally in the magnesia specimens. These have been identified as areas of carbon contamination. In such specimens, however, there is a marked contrast between the translucent and opaque regions.

Translucency occurs only in regions of high density (above approximately 99.6 percent of theoretical for BeO). The mottled appearance of many high-density hot-pressed samples is often due to the presence of regions of translucency in otherwise opaque material. A dramatic demonstration of the sudden onset of translucency occurs when powder is inadvertently loaded unevenly in the die. This load can result in one portion of the sample being higher in density than the other and, hence, produce translucency. This behavior is illustrated in Fig. 2 where three specimens are shown and where the density in all regions is greater than 98 percent for MgO, and 99.5 percent for both Al_2O_3 and BeO. The non-translucent areas are only fractionally lower in density.

Detailed studies of the effect of density on translucency of BeO were undertaken using a heavy liquid displacement technique. Small cores approximately

one in. in length and 1/8 in. in diameter were drilled from a block which showed alternating areas of whiteness and translucency. The grey samples were found to have a density greater than 99.6 percent, whereas the densities of the white samples ranged from 98.9 to 99.6 percent. These measurements preclude the possibility that greyness is due to graphite impurities.

B. MICROSTRUCTURE

Samples of the translucent oxides as well as high density white oxides were examined petrographically. The microstructure of all of these materials is characterized by essentially equant grains of the respective oxide which are unmodified by crystal edges. The only second phase observed was the porosity. Preferred orientation as a result of hot pressing was observed for the anisotropic oxides, Al_2O_3 and BeO .

A thin section was cut from a sample containing a lens-shaped region of opacity so that adjacent opaque and translucent areas could be studied. No sharp boundary existed between these zones at a magnification of 100X or greater, that is, the concentration of pores decreased gradually from the opaque area to the translucent area. Photomicrographs of these two regions are shown in Fig. 3. The translucent region is characterized by a relative lack of porosity as compared with the opaque material. The small amount of porosity that is present is almost entirely intragranular, that is, within the grain. The porosity in the case of the opaque sample was both intergranular and intragranular. Grain size was almost identical, ranging from about 30-60 μ . The preponderance of intragranular porosity in all specimens studies suggests that hot pressing is an effective means of removing grain boundary porosity.

Photomicrographs of translucent alumina and magnesia are shown in Fig. 4. The small amount of porosity which is seen in alumina is almost wholly intragranular. No pores were observed in magnesia which indicates that

any porosity which does exist is in the form of micropores. Grain sizes were 10-30 μ for alumina and below 10 μ for magnesia. The grain size of the translucent oxides, however, can be varied over a reasonably large range. Data reported by Richardson⁶ indicate that translucency is not affected by either the grain size or pore location in the finished sample.

C. THERMAL EXPANSION

The thermal expansion of the translucent oxides was measured in an air atmosphere to a temperature of 1000°C with the use of a Leitz dilatometer. The experimental data for the coefficient of expansion are given in Table 1.

Table 1. Coefficient of Expansion of the Translucent Oxides

Temperature, °C	Beryllia	Magnesia	Alumina	Sapphire
200	4.39×10^{-6}	9.65×10^{-6}	5.61×10^{-6}	5.25×10^{-6}
400	6.35×10^{-6}	11.65×10^{-6}	6.64×10^{-6}	6.63×10^{-6}
600	7.45×10^{-6}	12.28×10^{-6}	7.32×10^{-6}	7.35×10^{-6}
800	8.01×10^{-6}	12.78×10^{-6}	7.72×10^{-6}	7.64×10^{-6}
1000	8.57×10^{-6}	13.26×10^{-6}	8.09×10^{-6}	8.03×10^{-6}

A sample of sapphire cut such that the expansion measured was in the "c" direction was also measured for comparison. An experimental error was estimated to be $\pm 0.2 \times 10^{-6}$. Within this error there is excellent agreement between the measurements of the translucent alumina and those of the sapphire. The experimental data are in good agreement with values reported in the literature.^{7,8} No effect of high density was noted when these data were compared with expansion data taken on non-translucent bodies of slightly lower density.

D. THERMAL CONDUCTIVITY

At room temperature, heat is transmitted through ceramic bodies by conduction. Because the thermal conductivity of the gases in the pores is much

less than that of the bulk material, the thermal conductivity of a highly densified body, such as the translucent oxide, can be expected to be greater than that for less dense oxides. The effect of porosity on the thermal conductivity of alumina is shown in Fig. 5.⁹ The data, although preliminary, suggest that the dependence of the conductivity was greater than the theoretically predicted data, if the pores were assumed to be spherical.

The thermal conductivity of the hot pressed oxides measured at 100°C gave values of 0.48 for translucent beryllia,¹⁰ 0.06 for translucent magnesia,¹¹ and 0.06 for alumina⁹ near theoretical density. All values are given in cal/cm-°C-sec. Uncertainty in the experimental data reported in the literature prevents any reasonable comparison of these translucent oxides with other high density bodies. Of particular difficulty is beryllia whose erratic behavior prevented its inclusion in the porosity study previously referenced.⁹ Some preliminary data suggest that this may be due to a strong dependence of the conductivity on the crystallite orientation. In this case, the conductivity of the body would be a function of the degree of orientation introduced during fabrication.

At higher temperatures, thermal radiation through translucent bodies might be expected to make a significant contribution to the thermal conductivity predicted by Kingery and his co-workers.¹²⁻¹⁴ Such a contribution would result in the thermal conductivity reaching a minimum value and then exhibiting an increase. This behavior has been noted by several observers for opaque bodies, although the operative mechanism has been suggested to be non-radiative in nature.^{15, 16}

The thermal conductivity of translucent beryllia has been measured up to 2200°C.¹ The techniques which were used were similar to those previously reported.^{17, 18} The data were found to follow up to 1300°C the same curve previously reported for 99 percent dense beryllia: $1/K = 0.01875 t - 4.1$, where K is in cal/cm-sec-°C, and t is in °C.¹⁵ At temperatures above 1300°C, however, a departure from linearity was observed. In the case of

opaque beryllia of density about 99 percent of theoretical, this departure from linearity was observed only above 1800°C. At 1900°C, the thermal conductivity for the translucent material is 0.048 cal/cm-sec-°C, while for the opaque beryllia it was 0.036 cal/cm-sec-°C. The translucent sample, when heated to 2200°C, was found to lose its translucency. When re-measured, the data did not show the departure from linearity at 1300°C. This behavior strongly suggests that radiative heat transfer does make a significant contribution to the thermal conductivity of translucent materials at elevated temperatures.

E. MECHANICAL STRENGTH

It is well known that the strength of ceramic materials increases with increasing density.^{3, 19} In particular, the exponential relationship suggested by Ryshkewitch has found wide acceptance.^{20, 21} Knudsen examined much experimental and theoretical work and suggested that the strength is also dependent on grain size; that is, the strength increases with decreasing grain size.²²

The high density oxides, therefore, should exhibit higher strength than lower density materials provided that the grain size remained constant. For translucent materials, the degree of porosity is so low that the strength is, in all probability, dependent primarily on the grain size. Careful control of the pressure-temperature cycle during fabrication can control grain size to a certain extent in the translucent oxides.

While detailed studies on the mechanical properties have not been made to date, preliminary measurements on translucent beryllia of approximately 35-μ average grain size have repeatedly yielded values for the modulus of rupture from 35,000 to 37,000 psi. Consistent sets of specimens have not yet been measured at higher temperatures, and sample-to-sample variations in microstructure prevent a complete and unambiguous interpretation of results which have been obtained.¹

One preliminary series of modulus-of-rupture measurements, however, indicated that the tensile strength remained constant with temperature over the range 1000° to 1600°C , which was the highest temperature of measurement. The data are plotted in Fig. 6. Eight or more points were taken at each temperature. The average, plus and minus one standard deviation of the modulus of rupture, is shown. The data at 1400° to 1600°C were confirmed by a second series of samples, the strength of which in this temperature region fell within the same band.

It is probable that the entire set of measurements is too low by approximately 10,000 psi, most likely owing to large grain size and to inadequate care during machining. However, the achievement of transverse strengths of BeO in the region of 15,000 to 19,000 psi at temperatures above 1400°C is certain and is in marked contrast to earlier data.²³ In particular, the absence of a "knee" in the strength-temperature curve to 1600°C is noteworthy and parallels data on aluminum oxide at full density reported by Crandall.²⁴

F. ELASTIC PROPERTIES

The elastic properties of translucent hot pressed beryllia has been measured by Bente using a Sperry Ultrasonic Attenuation Comparator.²⁵ The experimental data are given in Table 2. Values are also shown for samples of lower density as well as for magnesia-doped material. The addition of the magnesia did not significantly change the elastic properties. The Young's Modulus and shear modulus were found to decrease with decreasing density. These values were lower than those reported by Ryshkewitch using a static test.²⁶

Table 2. Elastic Properties of Beryllia

Material	Density	Young's Modulus (psi $\times 10^{-6}$)	Shear Modulus (psi $\times 10^{-6}$)	Poisson's Ratio
BeO (translucent)	3.00	52.1	22.3	0.167
BeO (translucent)	3.00	53.6	22.8	0.177
BeO	2.93	51.4	21.9	0.173
BeO	2.93	51.4	21.6	0.190
BeO	2.82	49.2	20.7	0.188
BeO	2.82	49.1	20.6	0.193
BeO-MgO	2.98	53.4	22.9	0.165
BeO-MgO	2.98	53.3	22.3	0.195

G. DIELECTRIC PROPERTIES

The dielectric constant of the translucent oxides can be expected to be higher than that of less dense material. This phenomenon can best be explained by considering the ceramic to be a two-phase system, one of the phases being air with a dielectric constant of one. This system has been treated theoretically by several investigators.^{27, 28, 29} Some recent experimental data on the effect of porosity on the dielectric constant of alumina are shown in Fig. 7.³⁰ The experimental data are best described by the expression suggested by Bottcher.²⁸ At high densities the expression often attributed to Maxwell was also found to be applicable.^{29, 31}

The dielectric constant of the three translucent oxides has been measured at 1 mc with a Boston Research Company 260-A Q meter using a General Electric Hartshorn type 1690-A dielectric sample holder. The samples were in the form of discs, 2 in. in diameter and 0.25 in. thick. The dielectric

constant was found to be 10.5 for alumina, 10.4 for magnesia, and 7.2 for beryllia. The values for alumina and beryllia are in agreement with those of Newlan taken at 215 mcs, for a sintered translucent body, alumina (Lucalox), and a hot pressed translucent beryllia body prepared by the authors.³² These data were taken as a function of temperature up to 1200°C and are shown in Table 3.

Table 3. Dielectric Properties of Translucent Beryllia (BeO-MgO)

Temp °F	Loss Tangent		Dielectric Constant		
	220 mc	9375 mc	220 mc	9375 mc	2711 mc
RT	0.00025	0.00050	7.1	7.3	6.28
500	0.00030	0.00060	7.2	7.4	
1000	0.00025	0.00060	7.6	7.6	
1500	0.00060	0.00050	8.2	8.1	
2000	0.00055	0.00050	8.9	8.6	
2500	0.00550	0.00130	9.1	9.7	

H. OPTICAL PROPERTIES

The optical properties of the translucent oxides are of interest because of their potential use as high temperature irdomes. The infrared transmission is of special interest and has been measured between 1 and 200 μ . The samples that were studied were platelets, 0.75-in. square and 0.030 in. in thickness. Surfaces were machined using a 100-grit diamond surfacing wheel. The transmittance in the near infrared was measured using a Beckman Model IR4 infrared spectrometer. The experimental data are given in Fig. 8. In the visible range all of the samples were relatively transparent. They became opaque at longer wavelengths: about 5 μ for beryllia, 6 μ for alumina, and 9.5 μ for magnesia.

Measurements in the far infrared, above 25μ , were made using a Perkin-Elmer Model 301 double-pass instrument equipped with NaCl and KBr prisms. The experimental data are shown in Fig. 9. The beryllia began to transmit at about 30μ , the magnesia at about 40μ , and the alumina at about 50μ .

This behavior can be adequately explained by classical electromagnetic theory.³³ Most of the absorption in dielectrics, such as the oxides, is due to fundamental absorption since free electron absorption is negligible. The loss in transmission in the near infrared and the reappearance of transmission at longer wavelengths result from this fundamental absorption that is centered around the reststrahlen frequency. Measurements that have been made of the infrared transmission and reflectivity of a single crystal of beryllia and a translucent beryllia sample supplied by the authors indicate that the main reststrahlen frequency was about 730 cm^{-1} (14μ) with a subsidiary maximum on the high frequency side at about 1050 cm^{-1} (10μ).³⁴ No significant difference was noted between the single crystal and polycrystalline values.

The transmittance of the oxides should be quite sensitive to the porosity. Pores in a ceramic act as scattering centers. Since their size is large compared to the wavelength, such scattering is nonselective. Hence, the body appears white due to the scattered light. As the number of pores decrease the amount of light which is scattered also decreases until finally the material begins to transmit.

None of the translucent oxides appear to be completely transparent. In the case of alumina and beryllia, this transparency is reduced by reflection and subsequent scattering of light at grain boundaries. This arises from the fact that the index of refraction of the individual crystallites is dependent on the crystal orientation; in other words, the crystals exhibit birefringence. The birefringence value of beryllia is 0.014 and for alumina, 0.09.³⁵ Although the amount of light which is reflected is quite small, the number of grain boundaries is quite large. The result is that the ceramic has a "milky"

appearance. Due to its greater birefringence, beryllia should not be as translucent as alumina provided that the grain size is constant. Translucency should also increase with increasing grain size and become completely transparent when the grain size approaches the thickness of the test specimen. Magnesia, since it is cubic, does not exhibit birefringence and, hence, should be transparent. The absence of complete transparency of the magnesia is attributed to the presence of micropores.

IV. SUMMARY

Pure oxide powders of aluminum, beryllium, and magnesium were formed into dense translucent compact bodies by hot pressing.

The fabrication equipment consisted of double-action graphite dies inductively heated in a graphite tube furnace with pressure applied via two opposed uni-axial hydraulic cylinders. Temperature-pressure parameters for the Al_2O_3 were 1500°C at 6000 psi; for the BeO, 1750°C at 4000 psi; for the MgO, 1400°C at 6000 psi.

The densities of the translucent bodies were found to be more than 98 percent of theoretical for the MgO and more than 99.5 percent for the Al_2O_3 and the BeO. The color is a function of the density and neither of the grain size nor of the pore location. Translucent oxides are characterized by an extremely low porosity.

Thermal expansion was measured with a Leitz dilatometer. The data are in agreement with previously published values.

Data are presented on the high temperature thermal conductivity of BeO which indicate that there is a component of radiant heat transfer in dense oxides operative above 1300°C which increases with increasing transparency.

A series of modulus of rupture measurements are presented which indicate a straight line function of tensile strength with respect to temperature from 1000° to 1600°C .

The elastic properties of beryllia, including "opaque" material were measured at room temperature. Young's Modulus and the shear modulus decrease with decreasing density.

The dielectric constant for Al_2O_3 increases linearly with increasing density. The dielectric constant for translucent specimens of Al_2O_3 , BeO, and MgO were measured at 1 mc and found to be, respectively, 10.5, 7.2, and 10.4.

Transmission spectra in the near and far infrared region to 200μ have been measured for a 0.03 in. specimen of three translucent oxides. The approximate cutoff points in the shorter wavelength region is, respectively, 6μ , 5μ , and 9μ for Al_2O_3 , BeO , and MgO . Transmission begins again at about 50μ , 30μ , and 40μ for the specimens in the same order. The reflectance maxima (reststrahlen frequency) for BeO are at 10 and 14μ .

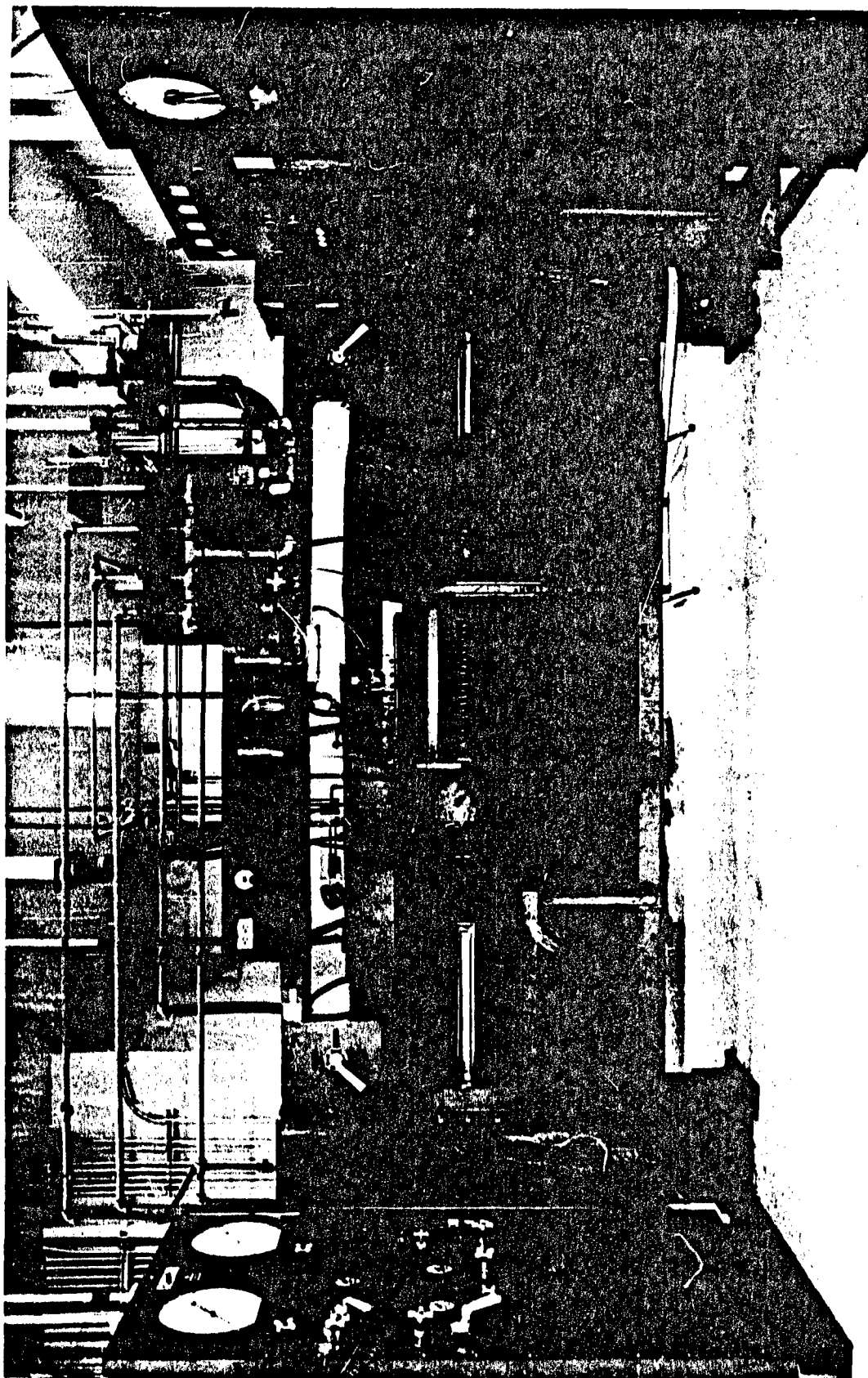


Fig. 1. Hot Press Equipment

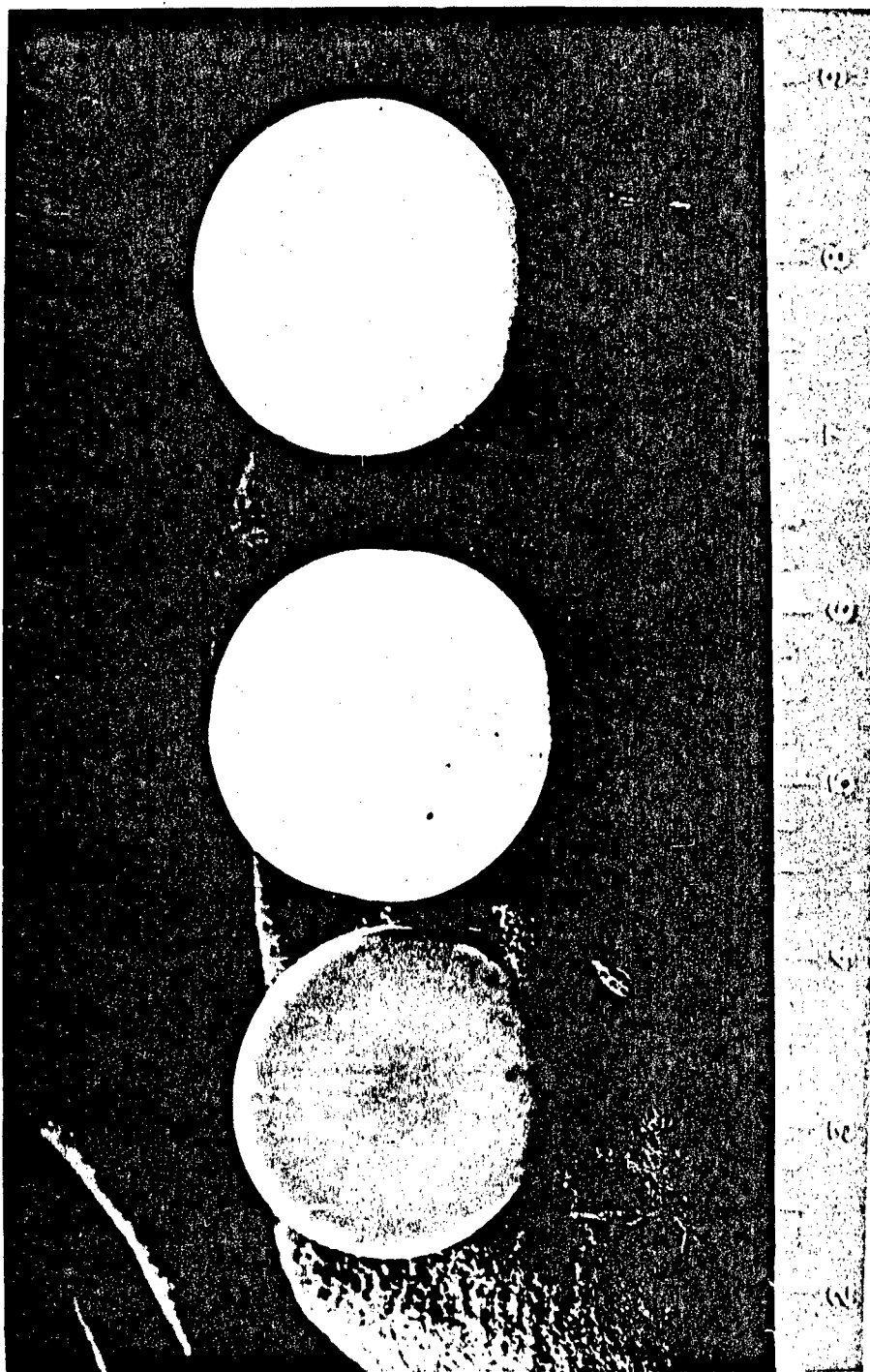


Fig. 2a. Translucent Oxides, Al_2O_3 , BeO , and MgO

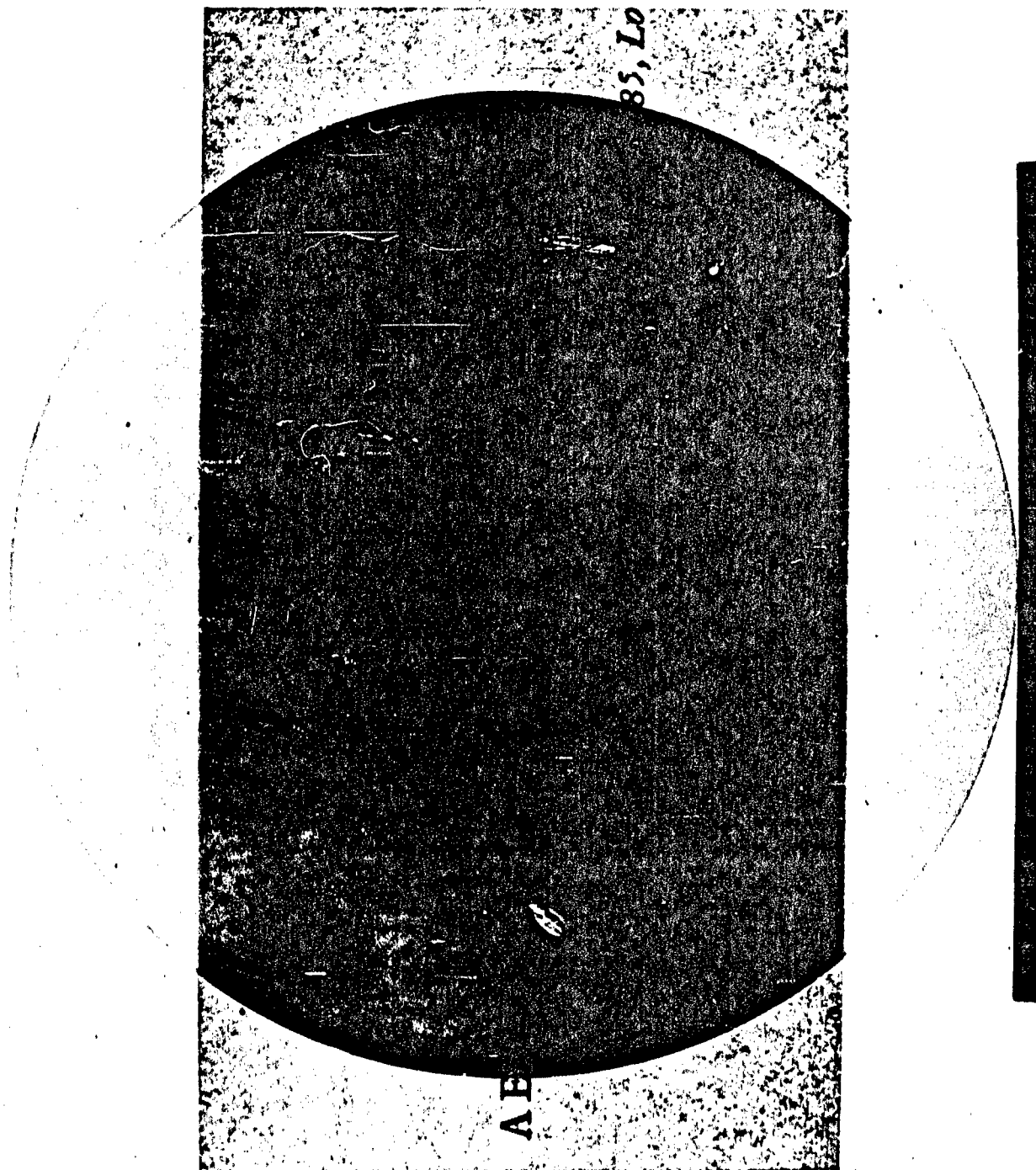


Fig. 2b. Translucent BeO



OPAQUE BeO



TRANSLUCENT BeO

Fig. 3. Photomicrograph of Hot Pressed BeO:
(a) Opaque; (b) Translucent



TRANSLUCENT Al₂O₃



TRANSLUCENT MgO

Fig. 4. Photomicrograph of Translucent Oxides: (a) Al₂O₃; (b) MgO

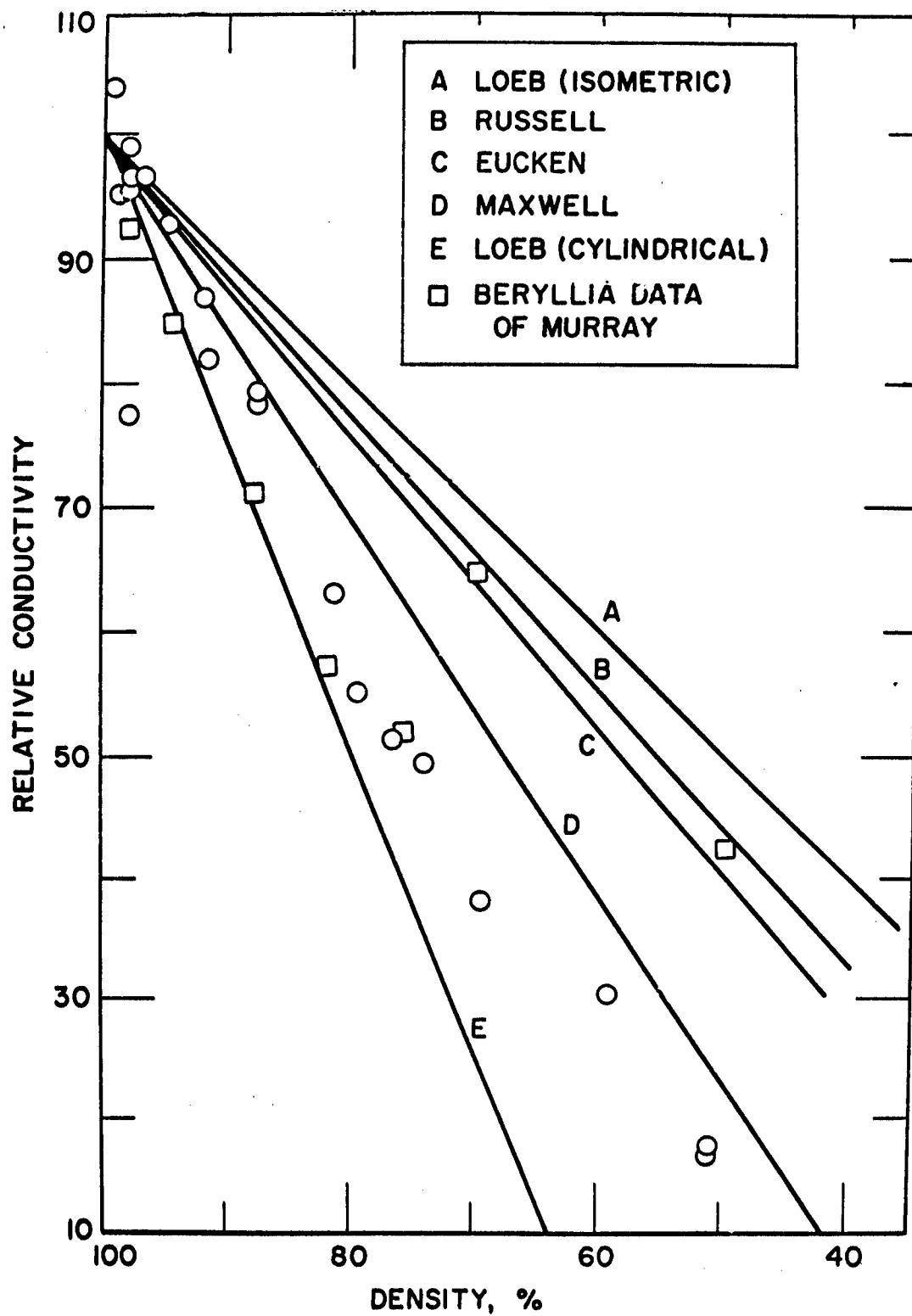


Fig. 5. Effect of Density on Thermal Conductivity of Alumina

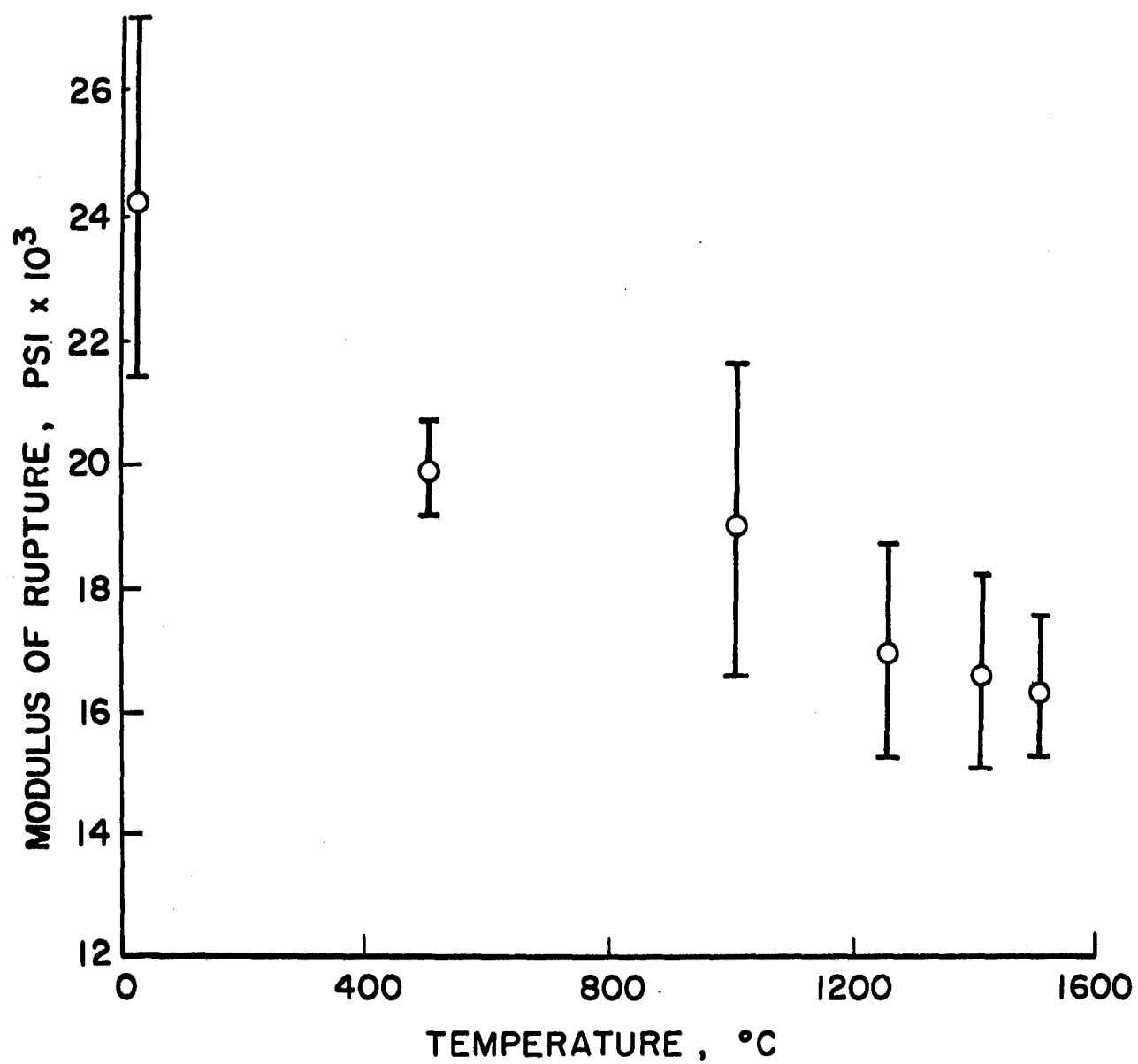


Fig. 6. Modulus of Rupture of BeO versus Temperature

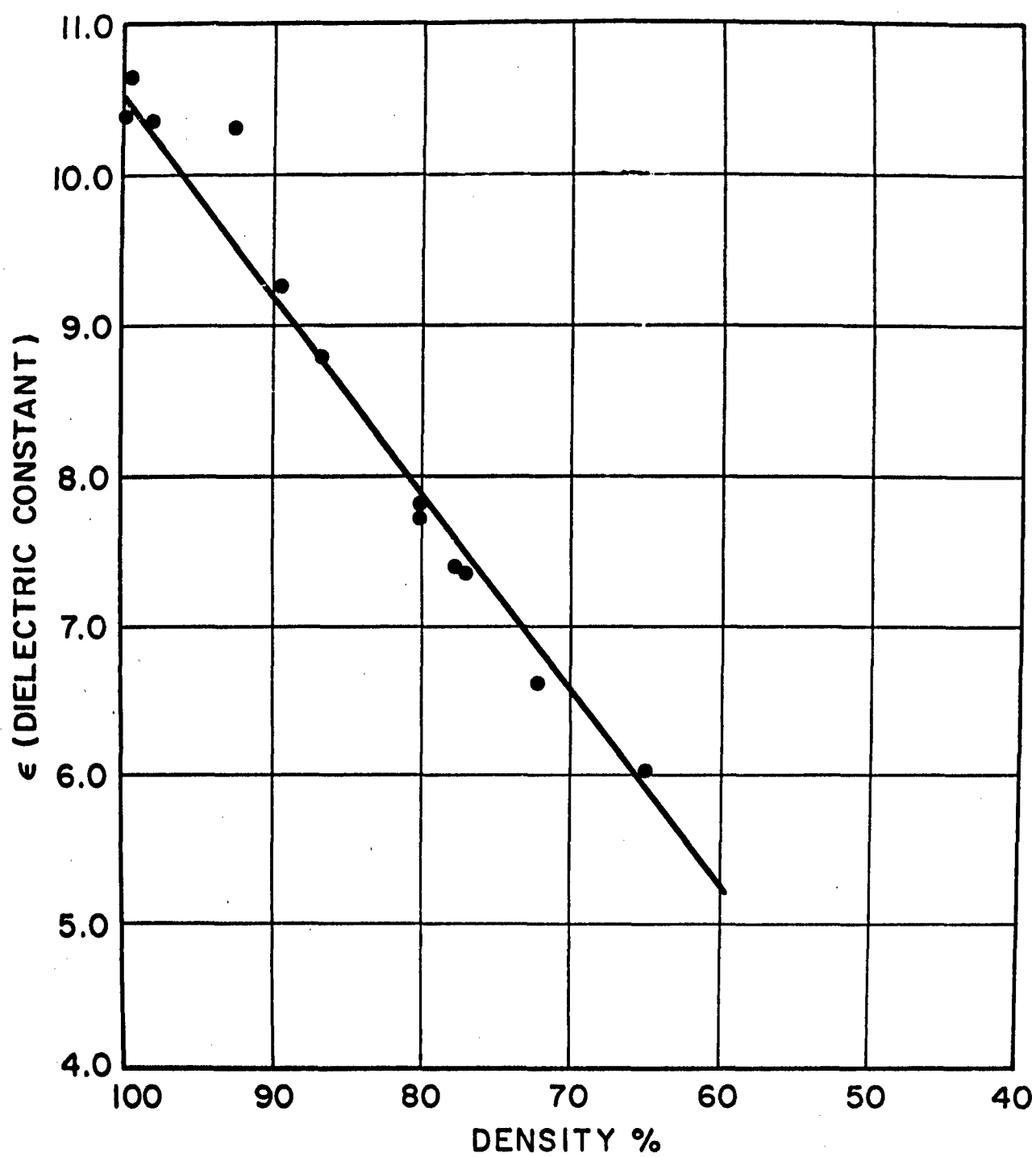


Fig. 7. Dielectric Constant versus Density for Al_2O_3

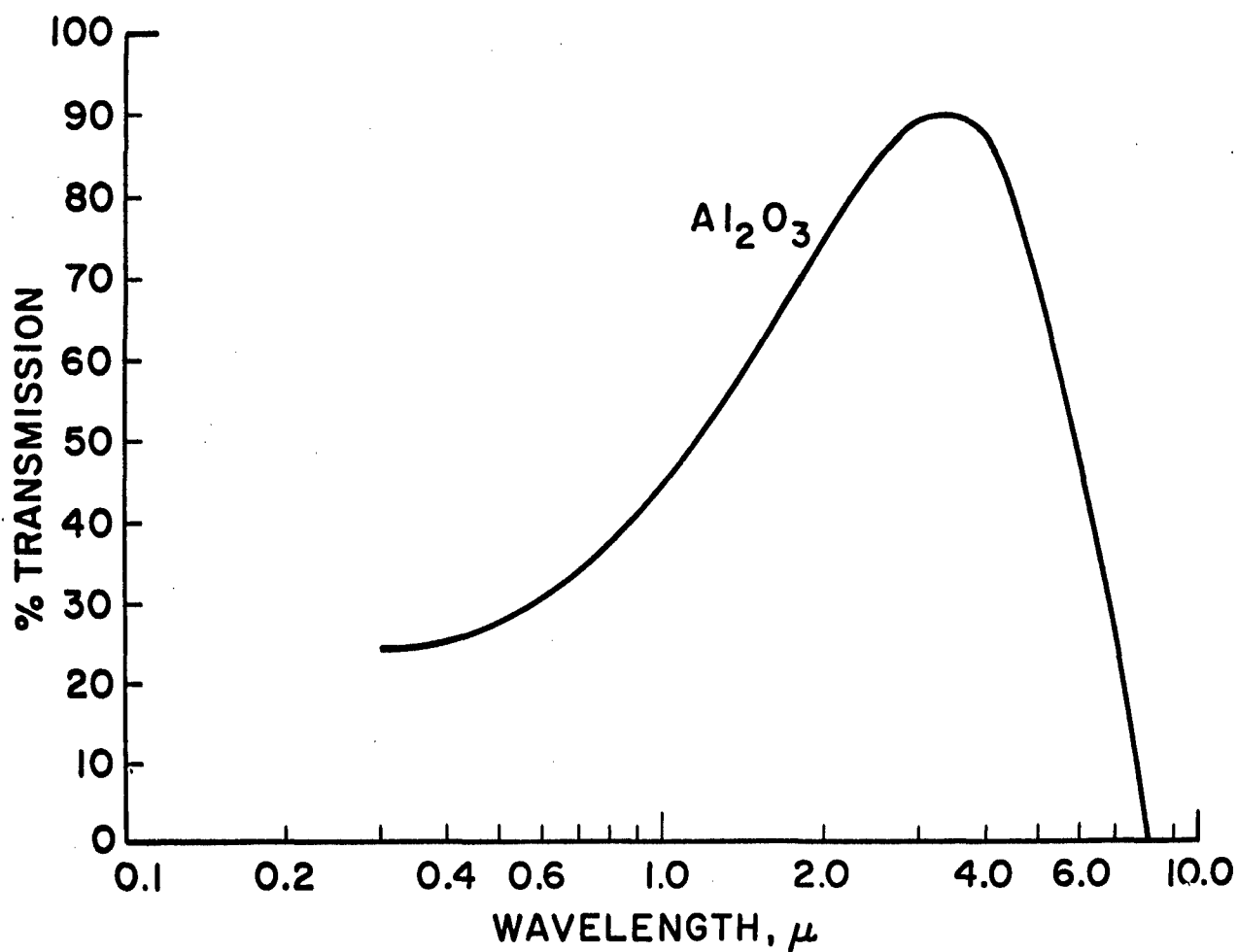


Fig. 8a. Near Infrared Transmission of Translucent Oxides (Al₂O₃)

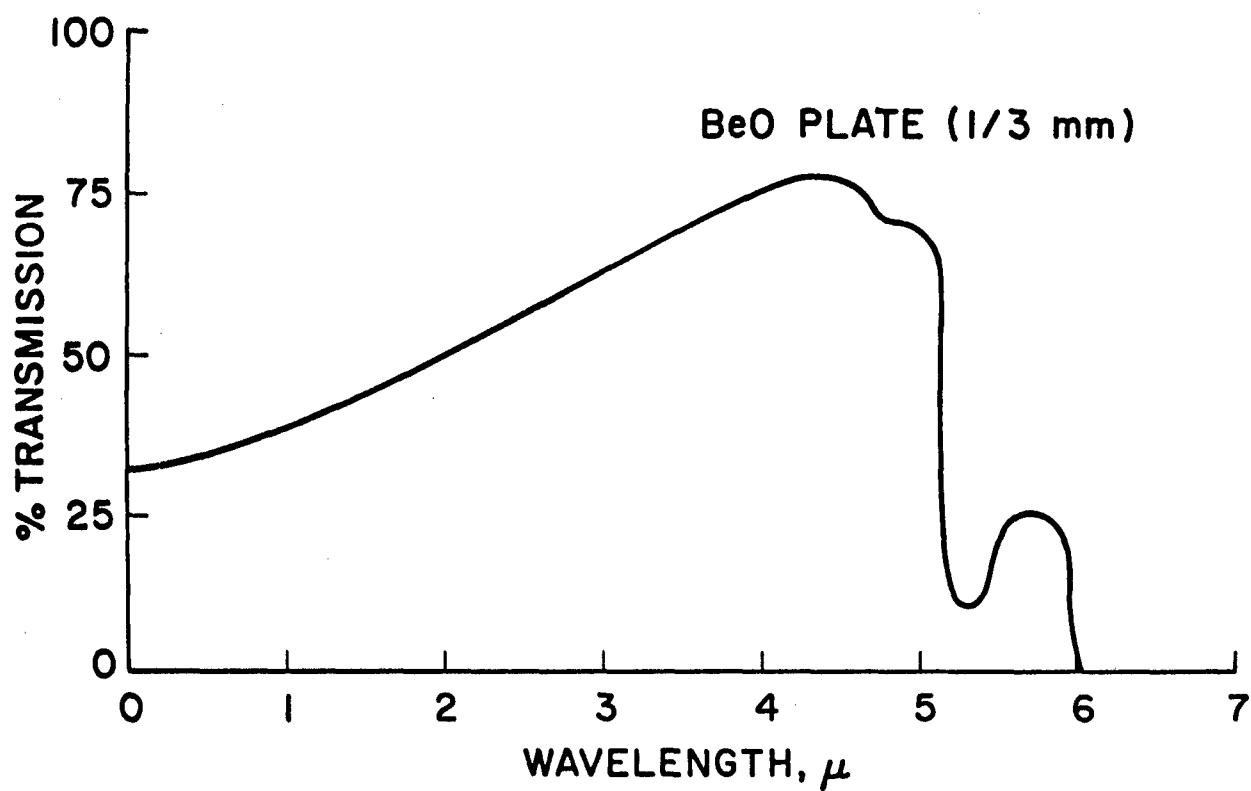


Fig. 8b. Near Infrared Transmission of Translucent Oxides (BeO)

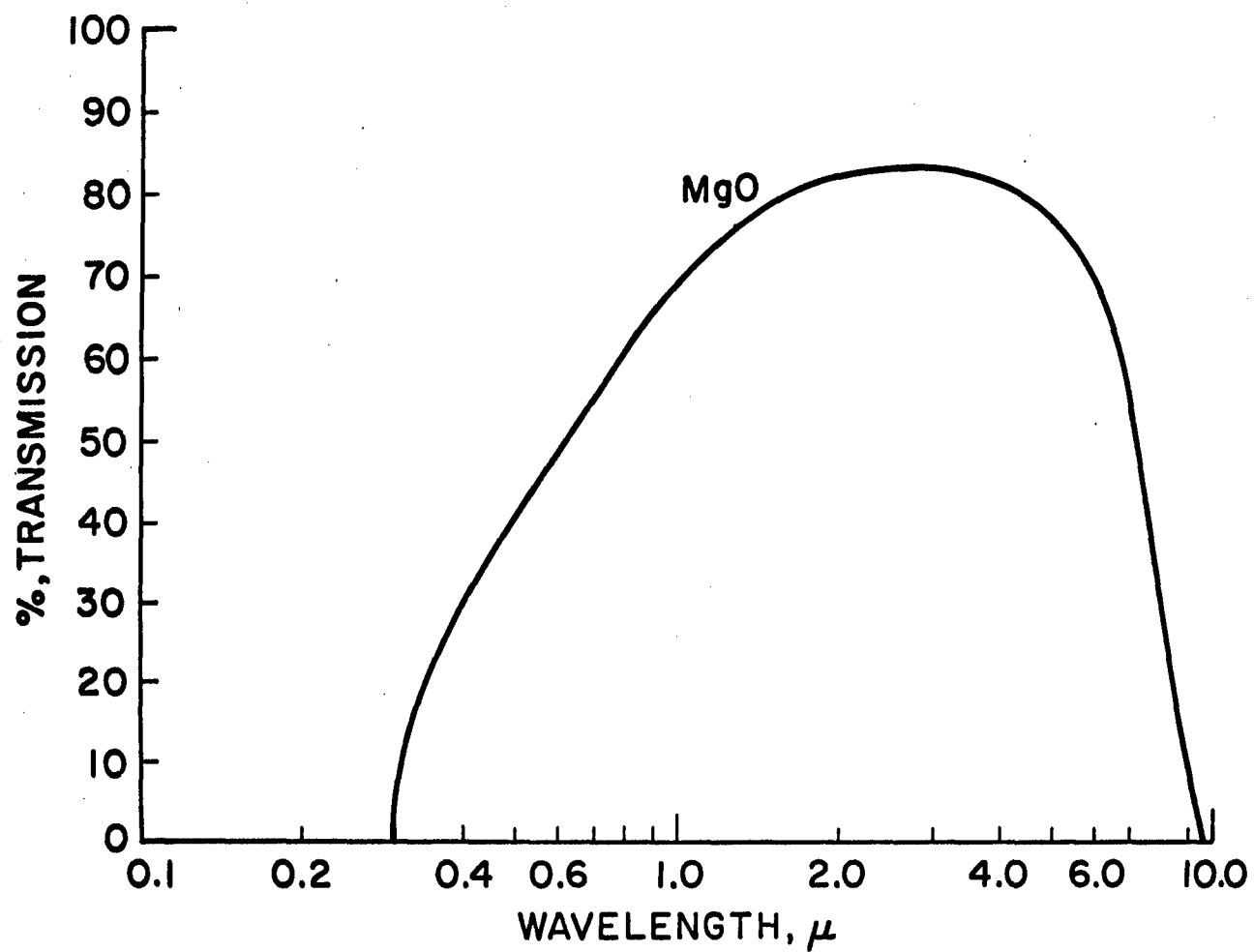


Fig. 8c. Near Infrared Transmission of Translucent Oxides (MgO)

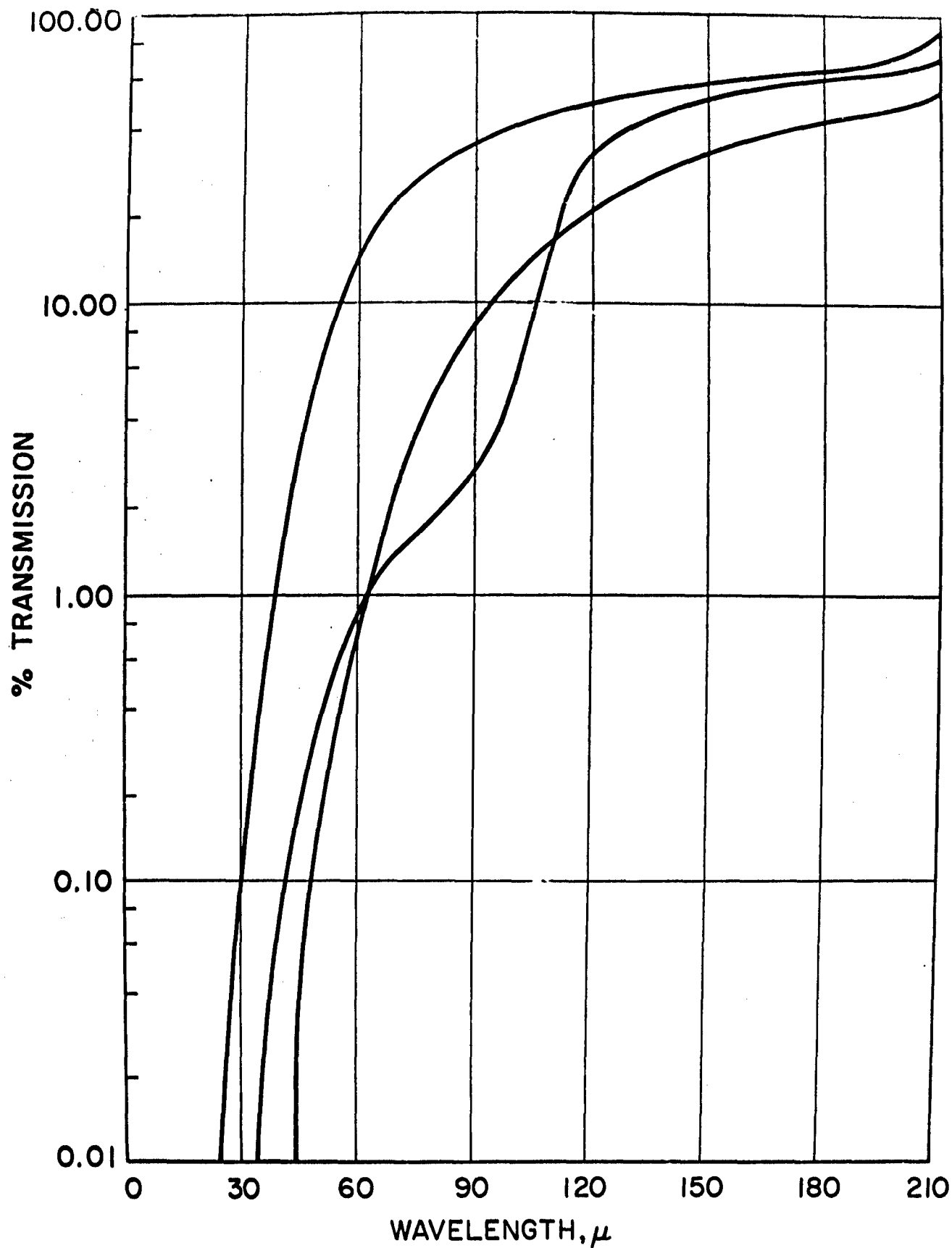


Fig. 9. Percent Transmission versus Wavelength for Al_2O_3 , BeO , and MgO

REFERENCES

1. J. D. McClelland, J. H. Richardson, and L. R. Franklin, "Fabrication and Properties of Translucent Beryllium Oxide", U. S. Atomic Energy Comm. , NAA-SR-6454, 13 pp (1961).
2. J. D. McClelland and E. H. Zehms, "End-Point Density of Hot Pressed Alumina", J. Am. Ceram. Soc. 46 (2), 77-80 (1963).
3. J. D. McClelland, "Kinetics of Hot Pressing of Ceramics", Powder Metallurgy - Proceedings of International Conference, New York, 1960, edited by W. Leczynski (Interscience Publishers, New York, 157-171, 1961).
4. C. A. Bruch, "Sintering Kinetics for the Lucalox Alumina Process", General Electric Research Laboratory, Report No. 61-RL-2778n (August 1961).
5. N. A. Weil, "Studies of the Brittle Behavior of Ceramics Materials", ASD-TR-61-628, 29-30 (April 1962).
6. J. H. Richardson and J. D. McClelland, "Preparation and Micro-structure of Translucent Beryllia", presented at the 14th Pacific Coast Regional Meeting of the American Ceramic Society, October, 1961.
7. C. J. Engberg and E. H. Zehms, "Thermal Expansion of Al_2O_3 , MgO , B_4C , SiC , and Ti Above $1000^{\circ}C$ ", J. Am. Ceram. Soc. 42 (6), 300-305 (1959).
8. A. Goldsmith, H. J. Hirshborn, and T. E. Waterman, "Thermophysical Properties of Solid Materials, Vol. III, Ceramics", WADC 58-476 (November 1960).
9. J. D. McClelland and L. O. Peterson, "Effect of Porosity on the Thermal Conductivity of Alumina", U. S. Atomic Energy Comm. , NAA-SR-6473 (1961).
10. R. K. Day, Private Communication.
11. "Physical Properties Brochure for High Density Magnesium Oxide," Minneapolis-Honeywell Regulator Company.
12. W. D. Kingery, et al., "Thermal Conductivity: X, Data for Several Prime Oxide Materials Corrected to Zero Porosity", J. Am. Ceram. Soc. 37 (2), 107-110 (1954).

13. F. Charvat and W. D. Kingery, "Thermal Conductivity: VIII, Effect of Microstructure on Conductivity of Single-Phase Ceramics", J. Am. Ceram. Soc. 40 (9), 306-15 (1957).
14. D. W. Lee, "Radiative Heat Transfer and the Conductivity of Ceramic Oxides", Mass. Instit. Technology, Thesis (1958).
15. R. E. Taylor, "Thermal Conductivity and Thermal Expansion of BeO at Elevated Temperatures", U. S. Atomic Energy Comm. NAA-SR-4905, (1960).
16. J. C. Jamieson and A. W. Lawson, "High Temperature Heat Conductivity of Some Metal Oxides", J. Appl. Phys. 29, 1313-14 (September 1958).
17. N. S. Rasor and J. D. McClelland, "Thermal Property Measurements at Very High Temperatures", Rev. Sci. Inst. 31 (6), 595-604 (1960).
18. J. D. McClelland and E. H. Zehms, "Thermal Conductivity of Magnesia from 1030°C to 1800°C", J. Am. Ceram. Soc. 43 (1), 54 (1960).
19. R. L. Coble, "Effect of Microstructure on the Mechanical Properties of Ceramic Materials", Section 22, Ceramic Fabrication Processes, W. D. Kingery (Ed.), (John Wiley and Sons, Inc., New York, 213-228, 1958).
20. E. Ryshkewitch, "Compression Strength of Porous Sintered Alumina and Zirconia, 9th Communication to Ceramography", J. Am. Ceram. Soc. 36 (2), 65-68 (1953).
21. W. Duckworth, Discussion on Ref. 20, J. Am. Ceram. Soc. 36 (2), 68 (1953).
22. F. P. Knudsen, "Dependence of Mechanical Strength of Brittle Polycrystalline Specimens on Porosity and Grain Size", J. Am. Ceram. Soc. 42 (8) 376-387 (1959).
23. S. C. Carniglia and J. E. Hove, "Fabrication and Properties of Dense Beryllium Oxide", U. S. Atomic Energy Comm., NAA-SR-5572, 15 pp. (1960).
24. W. B. Crandall, D. H. Chang, and T. J. Gray, "Mechanical Properties of Ultrafine Hot Pressed Alumina", Conference on Mechanical Properties of Engineering Ceramics, Raleigh, N. C., 7 March 1960. Proceedings published by Interscience Publishers, Inc., New York (1961).
25. G. G. Bentle, "Some Elastic Properties of BeO at Room Temperature", J. Nucl. Mater. 6 (3) 336-337 (1962).

26. E. Ryshkewitch, "Rigidity Modulus of Some Pure Oxide Bodies", J. Am. Ceram. Soc. 34, 322-326 (1951).
27. W. J. Brown, Jr., "Solid Mixture Permittivities", J. Chem. Phys. 23 (8), 1514-1517 (1955).
28. C. J. F. Bottcher, "Theory of Electric Polarization" (Elsevier Publishing Co., Houston, Texas, 1952.)
29. J. A. Reynolds and J. M. Hough, "Formulae for Dielectric Constant of Mixtures", Proc. Phys. Soc. LXX, 8-B, 769-76 (1957).
30. E. M. Cole, Aerospace Corporation Report to be published.
31. J. C. Maxwell, "A Treatise on Electricity and Magnetism", 3rd Edition (The Clarendon Press, Oxford, England, Vol. 1, p. 440, 1904).
32. B. M. Newlan, "High Temperature Dielectric Measurements in the VHF Range", Lockheed Aircraft Corp., Report No. LMSC 3-80-61-20, (December 1961) (NOrd-17017).
33. P. W. Kruse, L. P. McGlanchlin, and R. B. McQuistan, Elements of Infrared Technology (John Wiley and Sons, Inc., New York, 1962, 88-95).
34. J. R. Durig, et al., "Infrared Transmittance and Reflectance of Beryllium Oxide", J. Am. Optical Soc. 52 (9), 1078 (1962).
35. A. N. Winchell and H. Winchell, "Elements of Optical Mineralogy" (John Wiley and Sons, Inc., New York, 1959).

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